CONF-9509100--16

WHC-SA-2748-FP

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Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
Waste Management

Westinghouse
Hanford Company Richland, Washington

Hanford Operations and Engineering Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

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Date Published
May 1995

To Be Presented at
International Conference on
Nuclear Criticality Safety
American Nuclear Society
Albuquerque, New Mexico
September 17-22, 1995

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CRITICALITY SAFETY OF HIGH-LEVEL TANK WASTE

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ABSTRACT

Radioactive waste containing low concentrations of fissile isotopes is stored in underground storage tanks on the Hanford Site in Washington State. The goal of criticality safety is to ensure that this waste remains subcritical into the indefinite future without supervision. A large ratio of solids to plutonium provides an effective way of ensuring a low plutonium concentration. Since the first waste discharge, a program of audits and appraisals has ensured that operations are conducted according to limits and controls applied to them. In addition, a program of surveillance and characterization maintains watch over waste after discharge.

INTRODUCTION

High-level radioactive waste, a byproduct of the processing of nuclear fuels, is stored in underground storage tanks located in the high-level waste tank farms on the Hanford Site in Washington State. This waste contains low concentrations of fissile isotopes, primarily ^{235}U and ^{239}Pu, making it necessary to ensure that a nuclear criticality cannot occur. Criticality safety is based upon the assurance that this waste possesses characteristics which ensure subcriticality and there is no potential for significantly reducing the degree of subcriticality.

Storage tanks are located under at least eight feet of soil to provide shielding from radiation. Older single-shell tanks are inactive and no longer receive waste. Since 1971 waste has been sent to the newer double-shell tanks constructed of two steel shells separated by an annular air space. These tanks are 22.8 m (75 ft) in diameter with a nominal capacity of 3,800,000 L (1,000,000 gal). The outer shell is surrounded by reinforced concrete.

Tank waste is a complex mixture of chemicals. At least 49 waste streams have been routed to the tank farms since the first discharge in 1944, and many tanks contain waste from several streams mixed in various proportions.

The criticality safety philosophy governing waste storage requires that waste be discharged with a sufficiently large margin of safety to ensure that it remains subcritical into the indefinite future without supervision. Nevertheless, in addition to the many precautions taken before waste discharge to ensure safety, a program of surveillance and characterization maintains watch over the waste after discharge to provide additional assurance that there is no change in waste composition or distribution capable of compromising criticality safety.

Waste contains a very large ratio of solids to plutonium. Because it is very difficult, if not impossible, to separate the plutonium from these solids, they provide an effective way of controlling plutonium concentration. Stirring, mixing, pumping, or any other operation performed on the waste, tends to more completely mix the plutonium and solids.

USING A CONSERVATIVE MODEL OF WASTE COMPOSITION

Complexity and uncertainty in composition pose major difficulties in analyzing waste. This complexity and uncertainty makes it necessary to construct a model of waste designed to produce conservative results when used to calculate critical parameters. Conservative values, estimates, or assumptions are ones which produce "worse" (more limiting or severe in some way) results than the actual case. The intention is to make sure that, in spite of uncertainties, the real case is always as safe or safer than the model. Thus, if the model can be shown to be safe, the safety of the real situation has been shown.

Rogers [1] develops a conservative model based upon chemical analyses for 28 waste samples. Conservatism is assured by selecting the maximum concentrations of components that are good neutron scatterers and poor neutron absorbers, and minimum concentrations of components that are good neutron absorbers. The primary goal is to provide a waste composition known to have a smaller macroscopic absorption cross section than does any real waste. As appropriate, elements have been grouped together or even left out entirely. The resulting waste model provides a simple description which envelopes the many and varied samples from the individual tanks.

Subcritical parameters are calculated using the assumption of optimal water moderation and are independent of the actual content of water. No restrictions are placed on the water content of tank waste.

Table I shows the composition of solids used in the model. Water content is an independent variable and not part of the model. Since a low density of solids produces a low total neutron absorption cross section, the density of solids for the model is assumed to be 1,200 g/L, less than expected in real waste.

Table I. Composition of Conservative Waste Model.

Component	wt%
Oxygen (O)	40.7
Phosphorus (P)	6.9
Silicon (Si)	3.8
Sodium (Na)	21.5
Aluminum (Al)	7.2
Nitrogen (N)	0.0
Iron (Fe)	19.9

Although this model was developed to represent waste, it is not unique, and it may differ considerably for any given waste sample. A different composition having the same total absorption cross section should provide about the same critical parameters. This model should not be used for any purpose other than to provide a basis for determining conservative critical parameters for Hanford tank waste.

The model is conservative in comparison to individual waste stream. Combinations of waste types may be conservatively represented by the waste model, provided each individual waste type can be conservatively replaced by it. When this is done, any selected volume of real waste will contain a greater neutron absorption cross section than an equal volume of model waste. In this way, the entire contents of a storage tank can be seen to be conservatively represented by the model.

SAFETY PARAMETERS

Based upon the conservative waste model, the subcritical plutonium concentration limit is 2.6 g/L. If this plutonium concentration is not exceeded at any location in real settled waste solids, criticality is not possible, even if the configuration is not homogeneous.

Solids limit the ability of the plutonium to concentrate. Even after all liquids have been drained from the waste, the remaining solids ensure that the average plutonium concentration remains low.

When the solids density is divided by the subcritical plutonium concentration, a subcritical solids/plutonium mass ratio is found. For the model composition criticality is not possible when the solids/plutonium mass ratio exceeds 476. This subcritical limit applies to any waste for which the total neutron absorption cross section is greater than that for the waste model. This ratio is independent of waste density and its conservatism depends entirely on the low absorption cross section of the model components. When a subcritical limit is used as the basis for a discharge limit, it must be multiplied by a safety factor.

The subcritical mass ratio for waste composed entirely of natural uranium, aluminum, and/or silicon is larger than that of the model composition. The subcritical mass ratio of plutonium to natural uranium is 770. Whenever there is a possibility that the content of the above components will be high, either a subcritical limit of 770 or a more conservative safety factor should be used.

The Criticality Prevention Specification (CPS) for Hanford Site waste tanks limits the concentration of plutonium in discharged waste to not more than 0.033 g/L (0.125 g/gal). At the same time the solids/plutonium mass ratio in a batch prior to discharge must be shown to be at least 1,000. In reality, the actual solids content averaged over many discharges is expected to be much greater than this minimum permitted value.

A surveillance program periodically removes and analyzes samples taken from tanks. When tank inventory exceeds 10 kg plutonium, the tank-averaged solids/plutonium mass ratio is expected to be at least 5,000. In addition, the plutonium concentration found in the solids portion of a sample must be less than 1.0 g/L. If either of these conditions are not met, an investigation is undertaken to evaluate the quality of the measurement and the criticality safety of the tank.

CHARACTERIZATION OF TANK CONTENTS

Track Radioactive Components (TRAC) is a software program used to estimate waste composition in Hanford waste storage tanks. TRAC uses records of past waste additions and transfers to estimate the amounts and kinds of waste in each storage tank. Because of its limitations, TRAC-generated data is used conservatively in criticality safety evaluations.

Chemical analyses of waste samples are used to characterize waste. The highest quality analytical data available come from waste characterization reports for core samples. The great difficulty and expense in obtaining waste samples results in a database which does not fully describe the waste in tank storage. A characterization program is underway to increase our knowledge of waste composition.

Tank waste is maintained alkaline. Alkalinity aids in criticality safety evaluation by keeping the solubility of plutonium and other waste components low, thereby making them less mobile and inhibiting chemical processes which might change the composition.

Hobbs et al. [2] reports the upper limit on plutonium solubility in alkaline salt solutions representative of tank waste to be 0.017 g/L. Depending upon the makeup of the waste, the saturation concentration might be as low as 0.002 g/L. In practice, the low quantity of plutonium and the large quantity of intermixed solids work together to produce a very small concentration of dissolved plutonium.

The greatest plutonium concentrations are reported for samples composed of settled solids. From 312 solids samples reported by Braun et al. [3], the median plutonium concentration is 0.01 g/L, and the highest is 0.35 g/L. From among 306 liquid samples, the median plutonium concentration is about 0.0001 g/L. The two highest plutonium concentrations reported for liquid samples are just over 0.01 g/L.

Roetman et al. [4] establishes a conservative upper limit on the total plutonium in all Hanford Site waste storage tanks to be 981 kg (included is 45 kg of ^{233}U). Hanlon [5] states that the total volume of slurries and sludge is 62.3 million liters (16.4 million gallons). Based on the volume of slurries and sludge only, a conservative estimate of the average plutonium concentration at tank farms is found to be 0.016 g/L. The average solids/plutonium mass ratio is estimated to be about 74,500, which is about 150 times larger than required to assure subcriticality in homogeneous waste. At least one of the tanks may have a mass ratio as low as 10,000, but even this value provides a large margin of safety.

MECHANISMS THAT CHANGE PLUTONIUM CONCENTRATION

The goal of criticality safety is to ensure that tank waste will remain well subcritical for an indefinite time. Although discharged waste is well subcritical, conditions within the tank environment are not static, and the composition of the waste may change.

Unless the plutonium concentration increases above the minimum for which criticality is possible, the waste will remain subcritical indefinitely. An important concern of safety is the identification of processes capable of increasing plutonium concentration. There are two ways to achieve a high plutonium concentration: to receive waste with a high plutonium concentration or to concentrate plutonium after it reaches the storage tank.

Concentrating Mechanism: Exceeding Discharge Limits

Since the first waste discharge a program of auditing has been in place to overview compliance with process controls and guard against an abnormal discharge. Although it is theoretically possible that at some time in the past an abnormally large quantity of plutonium was discharged, the probability of such a discharge without detection is very small.

At a generating facility waste is accumulated in a holdup tank. Only after a batch accumulates is the waste discharged. Holdup tanks are potential locations for an accidental nuclear criticality and are therefore closely monitored to ensure compliance to criticality safety limits and controls. Audits are made and samples analyzed on a regular basis as part of a program to prevent abnormal accumulation of plutonium. If a violation is detected, the waste is not discharged to tank farms until after it has been returned to compliance.

Under normal conditions, the quantity of plutonium in a holdup tank is small, and it is mixed with a much larger quantity of solids. Accumulation of an abnormal quantity of plutonium, followed by its discharge to a storage tank, would require a failure of the auditing and sampling program. For the accumulation to be large, a major breakdown in both the processing operations and the auditing program would be required. For this plutonium to escape detection would require a failure of nuclear materials management. Multiple failures of operating and accounting procedures would have to go undetected before a large quantity of plutonium could be discharged to tank farms. Because the contents of holdup tanks are stirred, it is unlikely that plutonium would be concentrated in a small volume.

Concentrating Mechanism: Settling of Suspended Material

If left undisturbed, particulates suspended in a waste mixture will settle out. Settling is the most obvious, and also perhaps the most effective, mechanism for concentrating plutonium. The upper limit of the plutonium concentration in the settled layer is determined primarily by two factors: the areal density of plutonium and the quantity of solids present.

Settling probably accounts for most of the increased concentration found in analyzed samples, as compared to discharge limits. However, criticality is not possible unless the plutonium areal density exceeds 2.6 kg/m^2 (240 g/ft^2). To achieve this average areal density over the large area of a storage tank requires more than 1,000 kg of plutonium.

Concentrating Mechanism: Evaporation

Evaporation and settling produce waste configurations which are similar. Both processes may occur at the same time. Settled solids have a liquid layer covering them, but after evaporation, this layer may be absent. Solids in evaporated waste may achieve a lower water fraction than possible through settling alone.

Concentrating Mechanism: Chemical Separation of Components

Chemical processes in waste are often difficult to describe. Not only is the chemistry complex, there is a high degree of uncertainty in the chemical makeup.

Tank waste is maintained alkaline to prevent corrosion and to ensure that the plutonium remains combined with the solids. Alkalinity reduces the variety of chemical processes and permits dissolution of only a very low concentration of plutonium. Alkalinity simplifies characterization of the waste and simplifies criticality safety evaluation.

Natural conditions tend to prevent the accumulation of a large mass of plutonium in a small, compact volume. The majority of plutonium is precipitated and mixed with a large quantity of solids. These solids tend to prevent high plutonium concentrations and also shield the plutonium from the chemicals that would dissolve it. There is no mechanism to force precipitated plutonium into a localized region.

Even if it is assumed that chemicals dissolve and remove neutron-absorbing components, there are natural controls preventing criticality. The low plutonium areal density, the many different good absorbers present, and the natural tendency of components to disperse and mix are some natural controls. When the proportion of one particular absorber is decreased, it is likely that there will be several other absorbers unaffected. The large solids/plutonium mass ratio would continue to guarantee subcriticality. No chemical process has yet been identified that might credibly lead to criticality.

Dispersing Mechanism: Formation of Layers

An important mechanism of dispersal is the tendency of incoming waste to form layers. As a layer is formed, the waste is spread over a large area. Any localized volume of higher plutonium concentration within the incoming waste would be dispersed into a thin layer upon settling.

Dispersing Mechanism: Mixing

Mixing disperses plutonium and prevents accumulation. Although incoming waste is well subcritical, mixing increases the margin of safety even farther by dispersing the plutonium. No mechanism capable of increasing plutonium concentration appears capable of overcoming the dispersal and blending mechanisms.

THE IMPROBABILITY OF ACHIEVING CRITICALITY

The following is a generalized discussion of how the various mechanisms which influence criticality safety work together. Several independent events of low probability are required before a critical configuration is possible.

At the processing facility waste is accumulated in a large diameter, non-geometrically safe "holdup tank." Since an accidental criticality is possible in the holdup tank, it and the processes which produce waste are carefully monitored to ensure a low plutonium concentration and an adequate quantity of solids. No criticality has ever occurred in a holdup tank. This fact sets an upper limit on the quantity and concentration of plutonium discharged in a batch of waste.

The probability of criticality in a storage tank is much less than in a holdup tank. Once inside the much larger waste storage tank, a region of high plutonium concentration would be transformed into a thin layer and mixed with other waste. In addition, waste is discharged with an adequate quantity of solids to ensure subcriticality.

Most waste originated from processes governed by flowsheets for which every stage was carefully monitored and controlled. An abnormal condition of the waste would have been accompanied by a drop in the quantity or purity of the product plutonium and an investigation would have been undertaken to determine its cause. That a large quantity of plutonium, an extremely valuable material and the object of the entire production process, could have been improperly routed to waste without detection would be very difficult to explain.

For these reasons, it is very unlikely that abnormally large quantities of plutonium were ever sent to waste storage tanks. Multiple violations of process controls would be required to discharge a batch of highly reactive waste.

Most plutonium was sent to tank storage combined with a large proportion of depleted uranium. Separation of plutonium from the uranium is very unlikely, and criticality would not be possible unless the plutonium is separated. Therefore, for most waste, criticality would not be possible, regardless of procedural violations that might have occurred.

Nevertheless, the possibility must be considered that separated plutonium might be accidentally sent to tank farms. Before discharge it would be necessary to accumulate a region of extraordinarily high plutonium concentration. Upon discharge this would be transformed into a thin, flat region. Diffusion and circulation would spread the plutonium over a larger volume, thereby further reducing the concentration and the areal density.

If a second such batch were discharged, it would also be transformed into a thin, flat region. Regions of higher plutonium concentration should be randomly distributed. Even after many such abnormal discharges, the random distribution of the various regions of higher concentration would tend to produce a relatively uniform areal density over the large area of the storage tank.

Consider the hypothetical case in which an 18,900-L (5,000-gal) holdup tank is filled with waste containing plutonium at a concentration of 4 g/L. This extremely high concentration is hundreds of times greater than expected. Nevertheless, when this waste is discharged into the 22.9-m (75-ft) diameter storage tank, it would form a layer less than 5 cm (2 in.) thick. The areal density would be only 17 g/ft^2, as compared to the minimum critical areal density of 240 g/ft^2. Even after 10 more similar discharges, this waste would remain well subcritical. Neutron absorbers would ensure an even greater margin of safety.

If a scenario were to be selected to represent the sequence of events most likely to create a criticality configuration, it would probably be one in which incoming waste falls directly into deep supernatant liquid. Under certain conditions incoming waste might spread into a quasi-spherical volume before settling into a thin layer on the bottom. For criticality to occur it would be necessary for the concentration of suspended plutonium to exceed 7 g/L over a sizeable volume. At the same time the plutonium would have to be either dissolved or associated with very small particles capable of suspension.

To achieve the high plutonium concentration necessary for criticality requires either an incoming concentration exceeding 7 g/L or a buildup of suspended plutonium under the inlet pipe. In addition, the holdup tank would have to discharge more than a critical mass of plutonium, a quantity far greater than normal. A concentration of 7 g Pu/L would exceed by a factor of 200 the criticality prevention specification limit for the holdup tank. Because the holdup tank is stirred, such a region would be virtually impossible to create, unless the elevated concentration existed throughout the entire contents of the tank.

For a buildup of suspended plutonium to cause criticality, kilograms of plutonium would have to remain suspended within a limited volume while neutron absorbing solids settle to the bottom. Normally the suspended plutonium would be carried away from the inlet pipe by the outflow of incoming liquid.

For either situation, achieving criticality would require the simultaneous occurrence of several extremely unlikely events. Since criticality resulting from waste falling into supernatant liquid would occur within a short time following discharge, scenarios of this nature do not apply to waste already in storage tanks.

SURVEILLANCE PROGRAM

Since the beginning of fuel processing at Hanford, there has been a program of periodic audits and appraisals to ensure that all operations related to the production and disposal of radioactive wastes are conducted according to limits and controls applied to them.

In addition, a program of surveillance is conducted to monitor tank waste in the storage tanks. This program enhances the criticality safety program in two primary ways: it verifies that discharge criteria are met and it provides reliable information about waste. Analysis of characterization data provides assurance of the waste's high degree of subcriticality.

If a suspected nonconformance were to be detected, an investigation would be conducted to determine if the nonconformance is real or the result of measurement error. If the nonconformance is verified, a plan of recovery would be developed to return the system to compliance.

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